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On Energy Density in Dielectrics

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13. ABSTRACT (Maximum 200 words) Circuits for pulse sources to drive high power microwave (HPM) pulse generators require capacitors with high energy storage capability per volume. This paper explores various dielectric materials applicable to this requirement. Capacitors containing pyrogenically-grown amorphous silicon dioxide material are candidates for providing the desired capability.				
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ON ENERGY DENSITY IN DIELECTRICS

BACKGROUND

High power microwave (HPM) sources typically generate RF pulses of very high power and very short duration. RF pulses have been produced at power levels of 10 to 100 GW with pulse widths of approximately 100 ns [1]. Pulse power supplies drive these RF sources, and deliver hundreds of kilovolts and tens of thousands of amperes for about 1 μ s.

While the peak power of these devices is impressive, the total amount of energy residing in the RF pulse is quite modest: 1-10 kJ. Ten kilojoule is the equivalent of energy expended by a person to walk up a flight of stairs 10 m high. An average automobile battery stores 3000 kJ of energy. However, the driver of HPM sources must be able to release the energy in a very short time, which only capacitive or inductive storage elements can do. Unfortunately, the energy storage capability of capacitors is quite low. For this reason, the drivers of the HPM generators are usually very large and heavy.

ENERGY PER VOLUME STORABLE IN CAPACITORS

The Marx Generator is a capacitive, high voltage pulse generator, which is often used to drive HPM sources. It features a bank of capacitors that are charged in parallel and then switched in series by spark gaps to create the necessary high voltage pulse. Usually, these drivers are very voluminous and heavy in comparison to the actual HPM source. The size and volume of the drivers must be reduced to make these devices more practical. Therefore, a key feature is energy per volume that is stored in a capacitor.

The energy, W , stored in a capacitor is

$$W = 1/2 C V^2 \text{ joules} \quad (1)$$

where C is capacitance in farads, and V is the voltage to which the capacitor is charged.

$$C = \epsilon_0 \epsilon_r A/d \quad (2)$$

where ϵ_0 is permittivity of a vacuum = 8.854×10^{-12} farad/meter, ϵ_r is the relative (dimensionless) dielectric constant of the capacitor material, A is the area of the capacitor plates, and d their spacing.

The maximum voltage to which a capacitor can be charged depends on the "dielectric strength" S of the dielectric material. This is the maximum electric field strength the dielectric can stand before there is a breakdown. Hence, the maximum voltage V_{\max} is

$$V_{\max} = S \cdot d \quad (3)$$

Substituting (2) and (3) into (1), yields the maximum storable energy, W_{\max} , in the capacitor:

$$W_{\max} = 1/2 \epsilon_0 \epsilon_r S^2 A d \quad (4)$$

The volume Vol of the dielectric is $A d$. The maximum storable energy per volume W_{\max}/Vol is then

$$W_{\max}/Vol = 1/2 \epsilon_0 \epsilon_r S^2 \quad (5)$$

W_{\max}/Vol is a figure of merit for the energy storage capability of dielectric materials. It depends on the relative dielectric constant ϵ_r and the dielectric strength S . As shall be seen, the dielectric strength is the more important figure, because it appears as a factor squared. Equation (5) is not valid for ferro-dielectrics, whose dielectric constant depends on the field strength. For field strengths close to the dielectric strength, the dielectric constant of ferro-dielectrics can be a fraction of the constant for low fields. In this case, the stored energy is calculated:

$$dq/dV = C(V) = \epsilon_0 \epsilon_r(E) A / d \quad (6)$$

where dq is the differential charge and E is the electric field strength.

$$W = \int V dq \quad (7)$$

$$W = \int C(V) V dV \quad (8)$$

If the differential capacity $C(V)$ is known, then the integral can be evaluated numerically. $C(V)$ is the capacity that would be measured using an ac bridge with small amplitudes, with a dc bias voltage V at the capacitor. In the literature, Eq. (8) is sometimes given in the form

$$W/Volume = \epsilon_0 \int \epsilon_r(E) E dE = 1/2 \epsilon_0 \int \epsilon_r(E) d(E^2)$$

that is equivalent to (8). Here, E is the electric field strength in the material.

Table 1 lists the dielectric values and figures of merit discussed above for various materials.

Table 1 – Comparison of Dielectric Materials

Material	ϵ_r	S [kV/cm]	$\epsilon_r S^2$ [(kV/cm) ²]	W_{\max}/Vol [J/cm ³]
Teflon	2.1	400 - 800	3.4E5 - 1.3E6	0.015 - 0.058
Epoxy/Araldit	3.7	160	9.5E4	0.0042
Polyethylene	2.3	470	5.1E5	0.023
Ba-Titanate	1300	29	(1.1E6)	0.049
Polyethylene - Teraphtalate	3.2	1600	8.2E6	0.36
Ruby Mica	5.4	1500 - 2200	12E6 - 26E6	0.53 - 1.2
CPN17	7000	400	(1.1E9)	5.8
SiO ₂ Semi- crystalline	3.8	9800	3.6E8	16
SiO ₂ amorphous:				
[2]	3.9	3000		
[3]	3.8...3.9	5000-10000		
[4]	3.9	6000-9000		

DISCUSSION OF TABLE 1

Comparing energy storage per volume capability, barium titanate, a commonly used material, is better than polyethylene, but far inferior to mica. This results from the low dielectric strength of barium titanate in comparison to that of mica, despite the much higher dielectric constant of barium titanate. However, even if the dielectric strength of barium titanate could be improved through higher purity and different processing, the saturation of the ferro-dielectric would limit the improvement. Polyethylene terephthalate comes close to mica, but, at this time, the dielectric loss of this material at higher frequencies (10 MHz and above) is unknown. From the lower frequency data available, it is likely that the dielectric loss increases rapidly above 1 MHz.

CPN17 is a material which was developed by CERAM PHYSICS Inc., Westerville Ohio 43081, under AFOSR SBIR Phase II F49620-86-C0029 [5]. The composition of the material is $\text{PbO CdO Nb}_2\text{O}_5$. Its relative dielectric constant peaks to 7000 at 77°K. At field strengths close to the dielectric strength, the capacity of the material is only a fraction of that at low field strengths. Therefore, Eq. (5) is not valid for this case.

By far, amorphous silicon dioxide exhibits the highest energy storage per volume. It is superior to mica by a factor of 13 to 30 and to polyethylene by a factor of 700. However, the thickness of the dielectric was only 620 nm, as measured by a capacitive method. Measurement using an interferometer yielded a thickness of 735 nm.

MEASUREMENT OF SAMPLES OF AMORPHOUS SILICON DIOXIDE

Measurements were made on silicon dioxide-based capacitor samples that were fabricated in the Nanoelectric Processing Facility (NPF) of the Electronics Science and Technology Division (ESTD). The processing included RCA pre-oxidation cleaning on a 2 in. prime grade n-type <100> silicon substrate with 3×10^{15} phosphorus doping, pyrogenically thermal grown silicon dioxide at 1050°C for proper time, then evaporated the aluminum electrodes with thickness of 200 nm, 0.76 mm (30 mil) diameter, and 11.4 mm (45 mil) center to center spacing.

Using a capacitance bridge at 1000 Hz, the capacitance was measured as 25.5 pF. With a relative dielectric constant of 3.9 for silicon dioxide, the thickness of the dielectric was calculated to be 620 nm. Subsequently, a slowly rising dc voltage (5V/s) was applied to the capacitor through a 1M Ω current limiting resistor. Breakdown occurred at 600 V. Capacitance and breakdown voltage were consistent in several capacitors measured. Therefore, the dielectric strength was measured to be 9.7E6 V/cm.

The silicon dioxide was grown on a silicon substrate by oxidation. However, only very thin films can be grown this way, because the oxide layer prevents farther penetration of oxygen into the underlying silicon.

Commercial thermal-grown silicon dioxide is usually fabricated between 900°C and 1100°C. It is in the amorphous state as fused silica, or fused quartz. Quartz is the crystalline phase of silicon dioxide fabricated above 1710°C. Below that temperature the amorphous state exists but is thermodynamically unstable. A tendency of transformation from the amorphous to the crystalline state (devitrification) exists at temperatures below 1710°C.

Furthermore, pyrogenically-grown silicon dioxide capacitors with doped polysilicon electrodes were measured also. The fabrication procedures were similar to the aluminum sample with a 300 nm thick LPCVD (low pressure chemical vapor deposition) polysilicon layer doped with phosphorus at 900°C to yield a sheet resistance of 30 Ω /square, then photo-patterned square electrodes with a side length of 1.14 mm (45 mil). The capacitance of these capacitors was then measured to be 70 pF. The loss factor at 1 kHz was 0.26, which is quite high, and most likely caused by the high series resistance of the doped polysilicon material. Calculation of the thickness of the dielectric using the capacitance and the dimensions of the electrodes yielded a thickness of 640 nm. The dc breakdown voltage was measured as 120 V, yielding a dielectric strength of 1.9E6 V, which is only one fifth of that of the other sample.

CONCLUSION

It is known [6], [7] that, in general, the dielectric strength tends to decrease with increasing thickness of the sample. This was verified on crystals of rocksalt of varying thickness. The reasons proposed for this effect are inclusions of air pockets, impurities, stress and irregularities at crystal boundaries and, with increasing thickness, increasing free space length of accelerated electrons. It appears to be possible that if these problems could be avoided, a material with superior dielectric strength could be manufactured. In particular, if a single crystal of very high purity could be produced, there would be very few free electrons causing breakdown, and thus eliminating the reduction of dielectric strength with increasing thickness. Further, for a capacitor with a peak voltage capability of 100 kV, the thickness of the dielectric would only have to be 0.1 mm, which is quite thin. If this could be achieved, a new generation of capacitors with an increase of at least ten times the energy storage per volume could be manufactured.

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